OVERVIEW OF CURRENT COLLECTIVE PROTECTION FILTRATION TECHNOLOGY

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ABSTRACT

All collective protection filters consist of a particulate filtration stage followed by a vapor filtration stage. The particulate filtration stage consists of glass fiber high efficiency particulate air (HEPA) media providing greater than 99.97% filtration efficiency. The vapor filtration stage provides high capacity removal of all chemical warfare at efficiencies greater than 99.999%. Chemical vapors are removed by physical adsorption in the micropores of the activated carbon adsorbent as well as chemical reaction with impregnants deposited in the larger pore structure of the adsorbent. While current protection against toxic materials is very good, improvements are being developed.

1. INTRODUCTION

This paper summarizes the ability of Nuclear, Biological, Chemical (NBC) Filters to remove chemical/biological aerosols and vapors. NBC filtration technology has gradually evolved over the 80 years since the beginning of the modern era of chemical warfare in World War I. Over this time the filtration performance of NBC filters has been extensively studied and technology improvements have been implemented on an incremental basis. From these studies and filter development work, a large database is available on the performance of Nuclear, Biological, Chemical (NBC) filters in removing chemical warfare agents. In addition, some effort has also been expended in characterizing NBC filter performance in removing Toxic Industrial Materials (TIMs), but the existing database on TIM filtration contains many gaps. However, from the scientific principles that apply to the technologies used in NBC filters, one is able to confidently predict, in general qualitative terms, the effectiveness of NBC filters in providing filtration of many TIMs. Performance against some compounds is uncertain however, and recently work has been undertaken to close these TIM filtration data gaps.

NBC filtration systems consist of a particulate filter to remove liquid and solid phase toxic materials followed by a vapor filter to remove toxic chemical vapors. The particulate filtration stage consists of HEPA grade filter media. The vapor filtration stage consists of a bed of granular activated carbon which has been impregnated with reactive chemicals. The particulate filtration stage is located upstream of the vapor filtration stage so that volatile material released by trapped liquids and solids is removed by the vapor filter.

2. PARTICULATE FILTRATION

NBC filters use standard glass fiber high efficiency air (HEPA) media to filter solid particulates and liquid droplets. In this section, the words "particle" and "particulate" refer to liquid droplets as well as solid particulates. HEPA media consists of a nonwoven sheet composed of glass and polymeric fibers ranging in diameter from about 0.5 to 10 micrometer. The fibers remove particles from air by mechanical trapping that includes impaction, interception and diffusional processes.[1] Impaction refers to the process by which particles, because of their inertia, deviate from the air stream passing around the fibers and collide with a fiber. Interception refers to the process by which particles, when passing in the air stream around fibers, come in contact with a fiber. Diffusion refers to the process by which random Brownian motion of the particle, resulting from the movement of the air molecules, causes that particle to contact a fiber. Once fiber contact is made, small particles are retained by the fibers by weak electrostatic forces. (Larger particles are trapped by their inability to pass through the small openings between fibers in the mat.) The impaction and interception processes predominate for particles greater than about 0.2 micrometer in diameter, while the diffusional process is important for particles less than about 0.2 micrometer in diameter. Thus, at about 0.2 micrometer particle size, penetration through standard fibrous HEPA media tends to be greatest. Figure 1 graphically depicts the particle size ranges in which the three processes are important and their impact on defining the most penetrating particle size.[2] HEPA media is in fact defined as media providing at least 99.97% filtration efficiency at 0.3 micrometer particle size. (The 0.3 micrometer particle size is a carryover from a small error in early determinations of the most penetrating particle size.) Particles larger than about 0.2 micrometer are removed by HEPA grade media with efficiency greater than 99.97%, as are particles smaller than about 0.2 micrometer.

Typical chemical and biological particulate dispersions in air are in the 1 to 10 micron range, and standard HEPA media provides filtration efficiency of better than 99.9999% in that particle size range. This high level of filtration efficiency is adequate to provide protection against all chemical-biological threats.

The HEPA media is pleated to increase the area of the material used, thus decreasing the airflow velocity through the media. Low airflow velocity results in high filtration efficiency and low pressure drop. As is shown in Figure 2, the filtration efficiency of HEPA media increases greatly with decreasing airflow velocity.[2] Also, since pleating results in the airflow being directed over a larger area, the filter can remove much more particulate mass without airflow resistance increasing to an unacceptable level.

For the traditional HEPA media used in NBC filters, filtration efficiency tends to increase with loading of particulate on the media. This increase occurs because the spaces between fibers are reduced in size by the accumulated particulate. However, this accumulated material has the unwanted effect of increasing airflow resistance. Particulate filter change-out is generally based on the airflow resistance rising to unacceptable levels. At the time of change-out, the filtration efficiency of a particulate filter is typically higher than that provided when the filter was new. The particulate loading which requires filter change-out varies greatly from one filter design to another. Factors contributing to the variability in aerosol capacity for different filter designs are (1) the airflow velocity through the media (can be controlled by the extent of pleating), and (2) the margin that the airflow resistance of the filter when new is below the maximum allowable value. The nature of the particulate material also affects airflow resistance and thus filter change-out interval. An equivalent mass of a smaller particle size particulate collected by a filter results in more airflow resistance than does a larger particle size aerosol.[1] Particulates that are hygroscopic tend to result in greater airflow resistance because of clumping of the collected material.

Examples of chemicals removed by the particulate filtration stage of NBC filters include the tear gases and low volatility nerve agents such as VX. Biological agents, such as anthrax, as well as radioactive particulates are efficiently removed by the HEPA media in NBC filters.

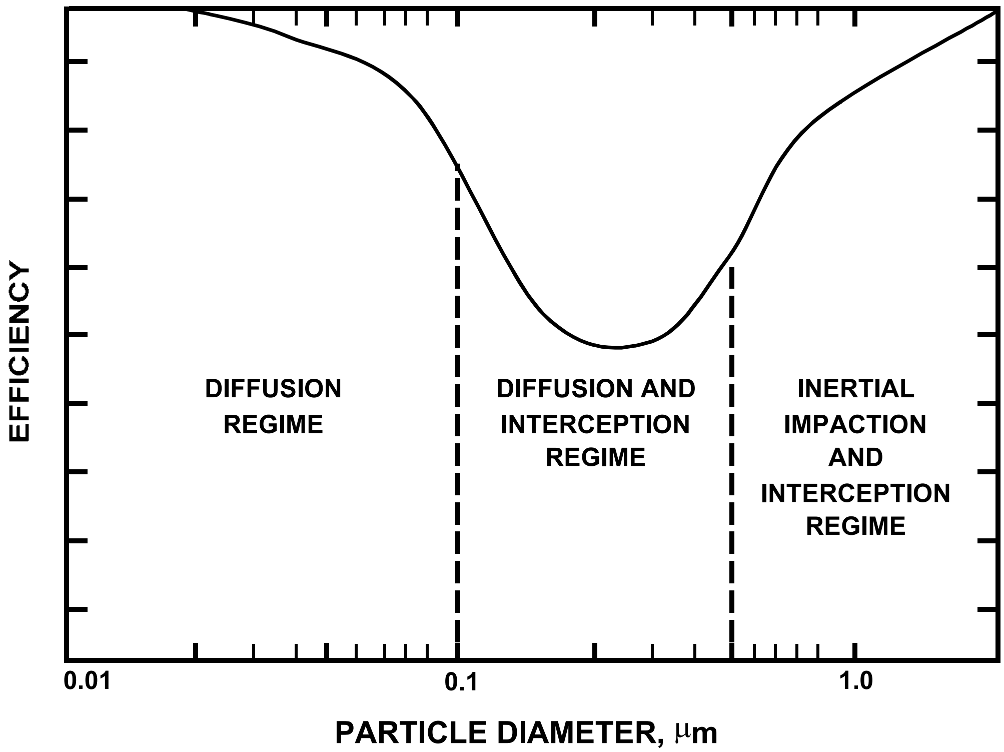

Figure 1. Particulate Filtration Processes [2].

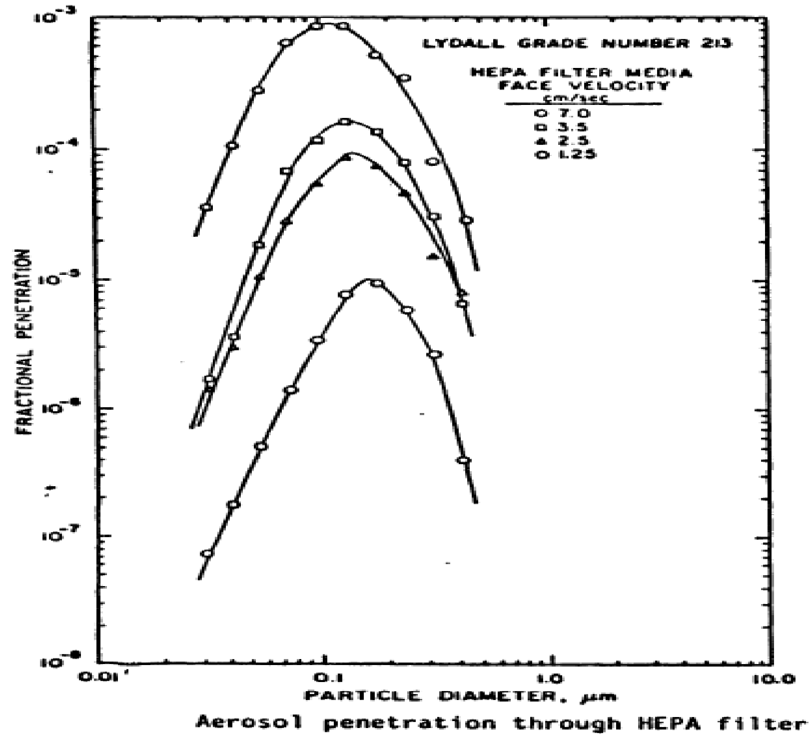

Figure 2. Filtration Efficiency as a Function of Particle Size and Airflow Velocity [2].

3. VAPOR FILTRATION

Immediately downstream of the particulate filter is the vapor filter consisting of activated carbon which has been impregnated with reactive materials. This impregnated, activated carbon filters vapors by two mechanisms, (1) physical adsorption in the pores of the activated carbon and (2) chemical reaction with the impregnants. Low vapor pressure chemicals, such as the nerve (e.g., GB) and vesicants agents (e.g., HD), are removed very effectively by physical adsorption in the microporous structure of the carbon. Relatively high vapor pressure agents, such as the blood agents cyanogen chloride (CK) and hydrogen cyanide (AC), are weakly physically adsorbed and will quickly penetrate a nonreactive activated carbon. Thus, specific reactive chemicals have been identified which chemically decompose high vapor pressure agents. These reactive chemicals are impregnated on the activated carbon to provide effective filtration of the complete spectrum of chemical warfare agents.

For approximately 50 years in the U.S., chemical warfare agent vapor filters contained the reactive adsorbent ASC Carbon, which is a coal-based activated carbon impregnated with chemical compounds containing copper, silver and chromium. Recently ASC Carbon was determined to be a hazardous waste under the Resource Conservation and Recovery Act (RCRA).[3] Because of high costs of disposing of spent ASC Carbon filters, ASZM-TEDA Carbon (a chromium-free carbon) was developed and has been adopted in nearly all U.S. NBC filters produced since 1993. The protection provided by ASC Carbon and ASZM-TEDA Carbon against chemical warfare agents is nearly equivalent.

ASZM-TEDA Carbon is a coal based activated carbon impregnated with copper, zinc, silver and molybdenum compounds, as well as with triethylenediamine (TEDA). ASZM-TEDA Carbon typically uses a special subgrade of BPL activated carbon (manufactured by Calgon Carbon Corporation) as the substrate for the impregnants. BPL Carbon is produced from bituminous coal and is highly activated to produce a surface area of about 1200 square meters per gram and a micropore volume of about 0.45 cubic centimeters per gram. The carbon has substantial meso- and macroporosity to provide for rapid internal mass transfer and to accommodate the application of the reactive impregnants. The impregnant loadings on ASZM-TEDA Carbon are:

Impregnant	Loading in Percent
Copper as cupric carbonate	5
Silver in elemental form	0.05
Zinc as zinc carbonate	5
Molybdenum as ammonium dimolybdate	2
Triethylenediamine	3

NBC filters containing ASZM-TEDA Carbon provide a high level of protection against all chemical warfare agent vapors listed in FM 3-9 entitled "Potential Military Chemical/Biological Agents and Compounds". With few exceptions the following minimum levels of filtration performance are provided by NBC filters at their rated flow:

Agent Class	Example Agent	Filtration Performance, Ct (mg-min/m^3)
Nerve	Isopropyl methylphosphonofluoridate (GB)	300,000
Vesicant	Bis-(2-chloroethyl) sulfide (HD)	300,000
Blood	Cyanogen chloride (CK)	80,000
Choking	Carbonyl chloride or phosgene (CG)	120,000

Ct is the challenge concentration to the filter integrated with respect to the time over which the challenge occurred. Thus, an example of a scenario that would result in a filter performance Ct of 300,000 mg-min/m^3 would be a GB challenge concentration of 100 mg/m^3 for 3,000 minutes.

ASZM-TEDA Carbon was developed specifically to filter chemical warfare agent vapors. However, the sorbent is also effective in filtering a wide variety of industrial chemical vapors. The vapor filtration performance of this sorbent depends primarily on two factors - the vapor pressure and the reaction chemistry of the chemical. Chemicals with low vapor pressures are effectively filtered by physical adsorption alone. As a general rule of thumb, chemicals with a vapor pressure below about 10 mm Hg (at the temperature of the filter) are very effectively removed by physical adsorption in pores of the activated carbon. As one considers higher and higher vapor pressure chemicals, the ability of the sorbent to remove chemical vapors by physical adsorption decreases. For these higher vapor pressure chemicals, reaction with the impregnants is necessary for effective filtration performance. Table 1 provides a summary of the filtration mechanisms for some chemical warfare agents. From these descriptions of agent removal mechanisms, the dual role of physical adsorption and chemical reaction in achieving filtration performance is apparent.

As a first level screen to estimate the effectiveness of ASZM-TEDA Carbon in filtering a specific vapor, one considers the vapor pressure of the chemical. If the vapor pressure is such that physical adsorption does not appear to be adequate to filter the chemical, reaction with the impregnants (or perhaps the carbon surface itself) could be a removal mechanism. By considering the chemical reaction properties of the chemical and the impregnants, an assessment can often establish the potential for removal of a chemical vapor by reaction with the impregnants. However, some risk exists in basing a determination of filtration performance based on expected reactivity. Reactivity, which is known to exist between two chemicals in a laboratory flask, may not be manifested to the same extent on the surface of activated carbon.

Of course, the most definitive determination of the ability of a filter to remove a chemical is to perform experimentation in the form a chemical challenge of the filter in question (frequently called a filter life test). Nearly equivalent data can be obtained by performing chemical vapor challenge testing of small beds of the adsorbent used in the filter in question. This simulated filter data can be obtained by either performing a test at the same bed depth and airflow velocity as those of the filter or by interpolating performance from a data set, typically a life-thickness plot. A life-thickness plot is a graphical model in which chemical vapor breakthrough times are plotted versus adsorbent bed depth for several airflow velocities. Figure 3 is a life-thickness plot of the filtration performance of ASZM-TEDA Carbon when challenged with the nerve agent simulant, dimethyl methylphosphonate (DMMP). The performance lines at each velocity are straight, which is typical of chemical vapors removed by physical adsorption. Figure 4 is a life-thickness plot of CK filtration performance of ASZM-TEDA Carbon. The filtration performance lines for CK are slightly curved indicating a significant chemical reaction rate effect.

Table 2 lists several of the more important collective protection gas filters, their primary system applications, their rated flow rates, their adsorbent bed depths and their superficial airflow velocities at their rated flow rates. The bed depths are all two inches or less so as to provide for low airflow resistance. The superficial airflow velocities are in the range of about 8 to 25 cm/sec. These airflow velocities are rather low to provide the filtration performance required from the relatively thin adsorbent beds and, again, to provide for low airflow resistance.

TABLE 1. Mechanisms of Agent Vapor Filtration by ASZM-TEDA Carbon

Agent	Filtration Mechanism
Nerve	Strong physical adsorption, generally followed by slow hydrolysis of the adsorbed agent.
Blister	Strong physical adsorption, generally followed by slow hydrolysis of the adsorbed agent.
Phosgene (choking agent)	Weak physical adsorption combined with agent decomposition effected by the impregnants. Phosgene hydrolyses to form hydrogen chloride and carbon dioxide. The hydrogen chloride reacts with the copper and zinc carbonate impregnants to form copper and zinc chlorides.[4]
Cyanogen Chloride (blood agent)	Weak physical adsorption combined with agent decomposition effected by the impregnants. Cyanogen chloride likely undergoes hydrolysis catalyzed by the triethylenediamine impregnant followed by removal of the acid breakdown products (hydrogen chloride and cyanic acid) by the copper and zinc carbonate impregnants. Some cyanic acid may hydrolyze to form carbon dioxide and ammonia.[4]
Hydrogen Cyanide (blood agent)	Weak physical adsorption combined with agent decomposition effected by the impregnants. Hydrogen cyanide reacts with the copper(+2) and zinc carbonate impregnants to form copper(+2) and zinc cyanides. The copper(+2) cyanide converts to cuprous cyanide and cyanogen.[5] The cyanogen reacts with the ammonium dimolybdate impregnant likely forming oxamide, which is strongly physically adsorbed by the activated carbon.
Arsine (blood agent)	Weak physical adsorption combined with agent decomposition effected by the impregnants. At low relative humidity, arsine is oxidized by copper(+2) to form arsenic trioxide (As_2O_3) and arsenic pentoxide (As_2O_5). At high relative humidity, arsine is catalytically oxidized by the silver impregnant to form arsenic oxides.[4]

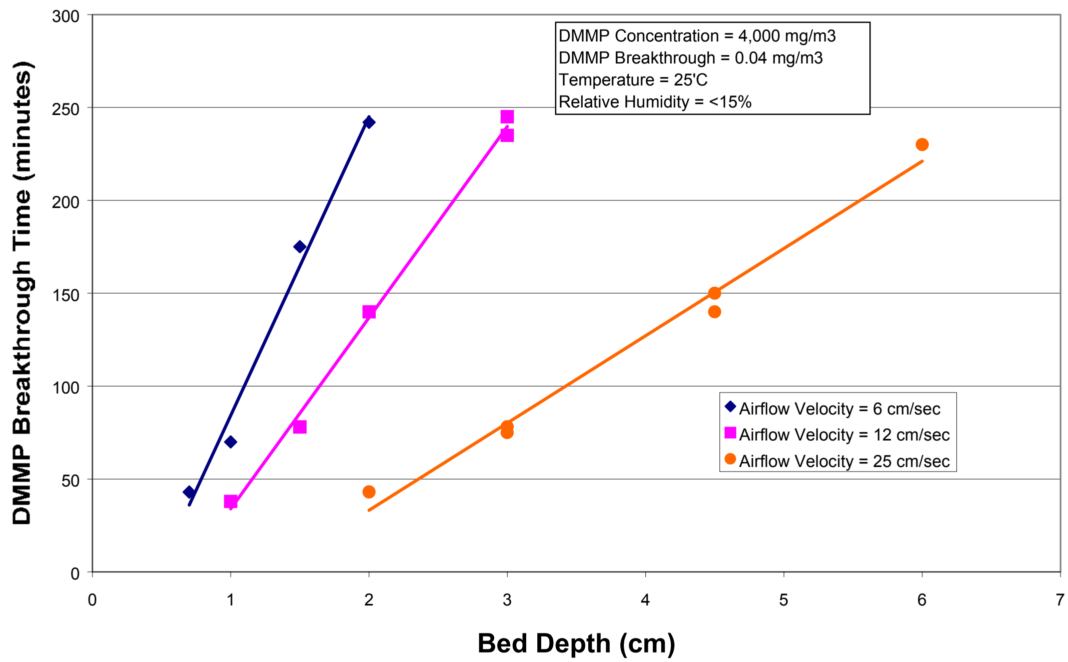

Figure 3. DMMP Life Thickness Plot for ASZM-TEDA Carbon.

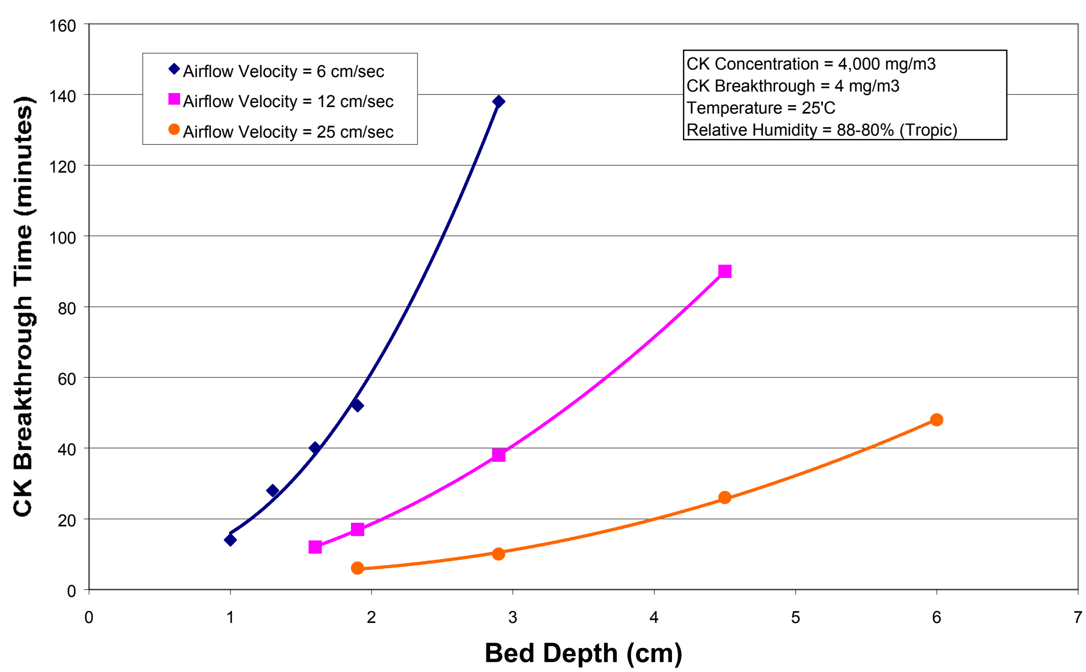

Figure 4. CK Life Thickness Plot for ASZM-TEDA Carbon.

TABLE 2. Collective Protection Gas Filter Characteristics

Filter	Application	Rated Flow (ft^3/min)	Flow Velocity (cm/sec)	Carbon Bed Depth (cm)
M12A3 Gas Filter (component in M8A3 Filter Unit)	Ventilated Facepiece for Armored Vehicle Crew	12	24.4	5.3
M18A1 Gas Filter (component in M13A1 Filter Unit)	Ventilated Facepiece for Armored Vehicle Crew	10	12.8*	3.4
M48 Gas-Particulate Filter	M1 Tank Overpressure and Shelters	100	22.9*	5.2
200 CFM Filter	Shelters & Ships	200	25.4*	5.6
M49 Gas Filter	Fixed Site	120 per tray	8.4	3.0

* Filter is radial flow. Airflow velocity is log-mean average for the carbon bed.

Even for chemicals which are removed very effectively by NBC filters, the amount of chemical that can be removed is limited by the space available for physical adsorption in the pore structure of the adsorbent and the amount of reactive impregnant contained on the adsorbent. In general, for every chemical vapor challenge to a filter, if continued long enough, breakthrough will eventually occur. (An exception to this statement would be for a purely catalytic reaction. However, such mechanisms are rare for filters operated at typical ambient temperatures.) The rate that the filter capacity is depleted is dependent on the concentration of challenge to the filter and the flow rate into the filter. Higher challenge concentrations and higher flow rates (such as heavier breathing rates for mask filters) result in shorter breakthrough times.

4. SPECIFIC NBC FILTER PERFORMANCE INFORMATION

NBC filters provide effective filtration against a broad spectrum of chemicals, but certainly not all chemicals. The chemical types described in this section are effectively filtered by NBC filters at a challenge level of at least 40,000 mg-min/m^3 (equivalent to a challenge concentration of 1,000 mg/m^3 for 40 minutes). The filtration efficiency for chemical vapors is generally at least 99.999%. This protection level is provided at all ambient temperatures and humidities except when near 100% relative humidity conditions exist. The filter must be free of liquid water to be effective. NBC filters provide good protection (per criteria above) against the following chemicals:

a. All chemical and biological warfare agents listed in FM 3-9 (Also known as NAVFAC P-467 and AFR 355-7).

b. All aerosols. Aerosol filtration efficiency is at least 99.97%, generally >99.9999% for typical threat aerosol size range of 1 to 10 micrometers. End of service life of the particulate filter is indicated by the airflow resistance rising to unacceptable levels.

c. Organic chemicals with vapor pressures of less than about 10 mm Hg at the temperature of the filter. However, consideration must also be given to the potential for reactivity of the chemical vapor with the impregnants on the carbon adsorbent in the filter or with the carbon surface itself. Toxic breakdown products could be produced should a reaction occur. If these toxic breakdown products are not effectively removed by the adsorbent, they could pose a hazard to the user of the filter.

d. The following acid gases:

hydrogen chloride
hydrogen fluoride
hydrogen bromide
fluorine
chlorine
hydrogen sulfide
sulfur dioxide
sulfuric acid

Limited protection is provided against chemicals with vapor pressures between about 10 and 100 mm Hg at the temperature of the filter. However, the filtration performance for chemicals of this vapor pressure range, especially against water insoluble chemical vapors, may be poor at high relative humidity. Also, although protection against chemicals falling within this vapor pressure range may be adequate for a period of time, the retention of the chemical by the filter is likely to be inadequate. On continued use of the filter, the chemical is likely to off-gas into the product air exiting the filter. NBC filters should not be relied on to provide a high level of protection against chemicals with vapor pressures falling in this range, but may be useful against brief, low concentration challenges. If exposed to toxic chemicals falling in this vapor pressure range, the filter should be replaced immediately after the chemical exposure ends.

NBC filters will not provide significant protection against the vapors of many chemicals with vapor pressures greater than about 100 mm Hg. Exceptions would be those organic chemicals which react with the impregnants or the carbon surface to produce breakdown products which are either nonhazardous or are retained by the adsorbent.

5. FILTER PERFORMANCE ASSESSMENT

5.1 Aerosol Filtration

Typically the filtration efficiency of an aerosol filter is measured by using a 0.3 micrometer monodisperse aerosol of either dioctylphthalate (DOP) or a polyalphaolefin known as Emery 3004 (also known as Ethylflow 164 or Durasyn 164). The filtration efficiency of the HEPA glass fiber media used in NBC filtration does not depend greatly on the physical state (liquid or solid) of the aerosol, or the properties of the aerosol other than particle size. Thus, the results of the DOP or Emery 3004 test are representative of the performance of the aerosol filter in removing chemical and biological agents. Filter testing with actual chemical or biological aerosols is rarely performed. Aerosol filtration efficiency testing is conducted during filter development and for production acceptance verification. In addition to efficiency testing, the extent that airflow resistance increases with loading is generally addressed during filter development using standard commercial test methods using materials such as “road dust”.

5.2 VAPOR FILTRATION

The approach to verify adequacy of vapor filtration is to inspect <u>adsorbent</u> performance through “gas life” tests with a variety of agents and to quantify <u>filter</u> performance with design limiting agents/simulants. For production lot acceptance of filters, testing includes only two design limiting agents/simulants (discussed below). However, during filter development, additional agent testing is generally conducted to verify the effectiveness of the filter design.

Before any production lot of filters is built, the Government verifies that the adsorbent production lot to be used in that filter production lot possesses the required level of agent filtration performance. As a result of this adsorbent testing, the ASZM-TEDA Carbon production lot to be used in the specific filter production lot is known to possess adequate agent sorption performance. Because it is economically impractical to test the adsorbent with every chemical warfare agent, only four difficult-to-remove agents have been selected to verify the adequacy of filtration performance of adsorbent production lots. The test agents/simulants used to verify adsorbent performance are cyanogen chloride, hydrogen cyanide, phosgene (carbonyl chloride) and dimethyl methylphosphonate (DMMP) as a simulant for GB. GB, usually through its simulant DMMP, is used to verify adsorbent performance because it is the most weakly adsorbed of those agents removed primarily by physically adsorption. Phosgene was selected to assess acid gas removal capability of the adsorbent since many agents possess acidic characteristics, either inherently or through hydrolysis of the agent. The two cyanide-containing agents (cyanogen chloride and hydrogen cyanide) are used in adsorbent performance verification because of their relatively low filtration capacity by the impregnant formulation of ASZM-TEDA Carbon. Thus, these two cyanide agents are important performance limiters for ASZM-TEDA Carbon. If a different adsorbent were used in NBC filters, a different set of design limiting agents may be required to adequately assess adsorbent performance.

Because the agent filtration performance of every adsorbent production lot is verified prior to its use, production lot filters need not be tested to verify filtration performance for the broad range of chemical warfare agents. However, the filter lot does need to be inspected to ensure that (1) a sufficient quantity of adsorbent has been used, (2) the adsorbent bed has been adequately packed, and (3) the reactivity of the impregnants on the adsorbent has not been compromised during filter production.

Verification of adequacy of the adsorbent fill quantity and packing is typically obtained through filter "protection life" testing using the nerve agent simulant DMMP (dimethyl methylphosphonate) augmented with some GB testing during filter development. Protection life testing is conducted by challenging the filter with a constant concentration of the chemical vapor in an airstream flowing at the rated flow of the filter under test. In some cases humidity is added to the airstream. GB is used because it is the filtration performance limiting agent representing all nerve agents and vesicants. Of those agents for which physical adsorption is the primary removal mechanism, GB has the highest vapor pressure. Physical adsorption by activated carbon, such as that used in NBC filters, is basically a phase change process in which the vapor pressure of the agent plays an important role. Ignoring for the moment the role played by the reactive impregnants on the adsorbent, the ability of a filter containing activated carbon to remove a specific chemical generally varies inversely with the vapor pressure of that chemical. Thus, GB is the most "weakly adsorbed" of those agents which are filtered effectively by physical adsorption alone. GB is therefore used as a basis to insure that military filters provide adequate protection against all known nerve agents and vesicants. In other words, if an NBC filter has the ability to remove agent GB to a certain extent, that filter will remove all other nerve agents and vesicants to an equivalent or greater extent. The simulant DMMP has been shown to effectively characterize the GB filtration performance of NBC filters and is frequently used as a substitute for GB to save money and avoid the toxicity hazard associated with agent testing.

Verification that the carbon impregnants have not been compromised during filter production is performed using cyanogen chloride (CK). CK has been selected for this role because, as mentioned above, it is one of the most difficult agents to filter adequately. In addition, CK removal is affected to the greatest extent of all agents by the effects of adsorbent "aging" resulting from exposure to high temperature and relative humidity. Generally, if an NBC filter can provide adequate CK protection, that filter generally will provide adequate filtration for all other agents for which chemical reaction is necessary for effective removal. During development of a new filtration system, a number of the "full-up” filters are tested for CK filtration performance. In addition, production lot acceptance testing is conducted with CK for filters with rated flows of 12 cubic feet per minute (CFM) or less. However, since costs associated with conducting high flow testing with a toxic compound are high, production lot acceptance CK testing of filters with rated flows greater than 12 CFM is not performed. Instead samples of carbon from the filter production line are CK tested in small tubes in a laboratory. These carbon samples have been exposed to the same environment as the carbon filling the filters on the production line. Thereby, assurance is obtained that the reactivity of the carbon has not been compromised by the filter production processes used by the manufacturer.

CONCLUSIONS

NBC filters provide a high level of protection against all chemical and biological warfare agents.

The current filtration technologies used in NBC filters are a pleated HEPA grade fibrous mat for aerosol filtration and activated carbon impregnated with reactive chemicals for vapor filtration.

NBC filters provide at least 99.97% filtration efficiency for submicron size particulates and at least 99.9999% filtration efficiency for particulates of micron size or greater.

The carbonaceous adsorbent used in NBC filters removes the vapors of low vapor pressure chemicals by physical adsorption in micropores, while the vapors of many higher vapor pressure chemicals are removed by chemical reaction with impregnants applied to the surface of the activated carbon.

NBC filters provide effective filtration against many toxic industrial materials, but some weaknesses do exist.

Adequacy of aerosol filtration performance is verified using a 0.3-micron aerosol of DOP or Emery 3004

Adequacy of vapor filtration is verified by means of extensive agent testing of the adsorbent, and by GB/DMMP protection life testing of the full-up filter.

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